

Thermoelectric Transport Coefficients for Massless Dirac Electrons in Quantum Limit

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We perform an analytic calculation of thermoelectric transport coefficients for massless Dirac electrons using the approach based on the Kubo–Středa formula and generalized Mott’s relation. The main focus of the letter is made on the properties of the Nernst coefficient in the vicinity of the Dirac point in quantum limit. We calculate magnetic field and temperature dependencies of the Nernst coefficient and compare our results with recent experiments in α -(BEDT-TTF)₂I₃ organic conductor. We argue that the Zeeman splitting is important to understand the experimental data at high magnetic fields.

KEYWORDS: Dirac electrons, Dirac point, thermoelectric coefficients, graphene, α -(BEDT-TTF)₂I₃, Nernst effect, generalized Mott’s formula

Unusual thermoelectric properties of graphene have attracted considerable interest. In graphene conducting electrons can be described by a Weyl equation that in quantizing magnetic field leads to relativistic Landau levels with energies $\pm \hbar \omega_c \sqrt{n}$ ($n = 0, 1, \dots$) with ω_c and \hbar being the cyclotron frequency and Planck’s constant respectively. The important difference of relativistic Landau levels from the non-relativistic case is the existence of $n = 0$ level with zero energy. In quantizing magnetic field, when the chemical potential is close to $n = 0$ Landau level, Seebeck and Nernst coefficients show an anomalous behaviour.^{1,2}

Massless Dirac fermions were also experimentally found in α -(BEDT-TTF)₂I₃ organic conductor under pressure.^{3,4} Tight binding model^{5,6} and band structure calculations^{7,8} revealed the existence of the Dirac point in this material with conducting electrons obeying the tilted Weyl equation. Recently, an anomalously large Nernst signal was reported in this system at high magnetic fields..⁹

Recently, the Nernst effect was intensively studied for massless Dirac fermions in graphene^{10–12} and on the surface of a topological insulator..¹³ Various theoretical approaches showed that the Nernst coefficient is greatly enhanced and can considerably exceed the Seebeck coefficient when the chemical potential is close to the Dirac point.^{10–12} This behaviour is sharply contrasted from the case with finite chemical potential in which the behaviour of the the transport coefficients is consistent with previous theoretical predictions for the non-relativistic two-dimensional electron gas.^{14,15}

From a theoretical viewpoint the calculation of the Nernst coefficient is a rather challenging problem since the standard linear response approach, based on the Kubo formula, gives unphysical divergence at zero temperature. Corrections arising from the thermal magnetization should be taken into account.^{16,17} Now it is well established that for the case of non-interacting electrons

the thermopower tensor satisfies the generalized Mott’s relation^{16,17}

$$\mathbf{S} = \frac{\boldsymbol{\sigma}^{-1}(T, \mu)}{eT} \int_{-\infty}^{\infty} d\epsilon f'(\epsilon) (\epsilon - \mu) \boldsymbol{\sigma}(0, \epsilon). \quad (1)$$

where T is temperature, $e > 0$ is an electron charge, μ is chemical potential, $\boldsymbol{\sigma}$ is conductivity tensor, $f(\epsilon) = \{1 + \exp[(\epsilon - \mu)/k_B T]\}^{-1}$ is Fermi–Dirac distribution function, f' denotes the derivative with respect to ϵ , and k_B is Boltzmann constant. The important property of the Mott’s formula is that, to calculate the thermopower tensor, one only needs to know the conductivity at $T = 0$ as a function of μ , $\boldsymbol{\sigma}(0, \mu)$.

The present letter is mainly devoted to the properties of the Nernst coefficient of massless Dirac fermions in the vicinity of the Dirac point in quantum limit where the distance between Landau levels is greater than temperature and impurity broadening. For this purpose we perform an analytic calculation of thermoelectric coefficients using the Kubo–Středa formula for conductivity and generalized Mott’s relation. We ignore the possible tilting of the Dirac cone and consider a simple case of energy-independent damping Γ due to the impurity scattering. In the limiting case when Γ is much less than $k_B T$ and $\hbar \omega_c$, we obtain an analytical expressions for Seebeck and Nernst coefficients. We show that the magnetic field dependence of the Nernst coefficient in the presence of the Zeeman splitting is different in $\Gamma \ll k_B T$ and $\Gamma \geq k_B T$ regimes.

In order to obtain the conductivity, $\boldsymbol{\sigma}(T, \mu)$, we consider a system of free massless Dirac electrons confined to a two-dimensional (x, y) -plane moving in a magnetic field B perpendicular to the plane. The model Hamiltonian is given by

$$H = -v_F \sum_{i=x,y} \sigma_i [-i\hbar \partial_i + eA_i(\mathbf{r})] \quad (2)$$

where v_F is Fermi velocity, σ_i is Pauli matrix, ∂_i denotes a derivative with respect to $i = x, y$, and $\mathbf{A}(\mathbf{r}) =$

$(-By, 0, 0)$ is a magnetic vector potential in the Landau gauge.

The solution of the eigenvalue problem for the Hamiltonian (2) leads to relativistic Landau levels $E_{n\alpha} = \alpha\hbar\omega_c\sqrt{n}$ ($n = 0, 1, \dots$) where $\alpha = \pm 1$ is the band index and the cyclotron frequency is given by $\omega_c = \sqrt{2}v_F/l_B$ with $l_B = \sqrt{\hbar/eB}$ being the magnetic length. The corresponding eigenfunctions are

$$\psi_{k0}(x, y) = \frac{e^{ikx}}{\sqrt{l_B L}} \begin{pmatrix} 0 \\ \phi_0\left(\frac{y}{l_B} - kl_B\right) \end{pmatrix} \quad (3)$$

and

$$\psi_{kn\alpha}(x, y) = \frac{e^{ikx}}{\sqrt{2l_B L}} \begin{pmatrix} \phi_{n-1}\left(\frac{y}{l_B} - kl_B\right) \\ \alpha\phi_n\left(\frac{y}{l_B} - kl_B\right) \end{pmatrix} \quad (4)$$

for $n = 1, 2, \dots$, where ϕ_n are the eigenfunctions of a harmonic oscillator. We imply periodic boundary conditions in the x -direction with L and k being the system size in the x -direction and the wave number respectively.

For non-interacting electrons the conductivity can be calculated using the Kubo–Středa formula¹⁸

$$\sigma_{ij} = \frac{ie^2\hbar}{2\pi} \int_{-\infty}^{\infty} d\epsilon f(\epsilon) \times \text{Tr} \left[v_i \frac{dG^+(\epsilon)}{d\epsilon} v_j A(\epsilon) - v_i A(\epsilon) v_j \frac{dG^-(\epsilon)}{d\epsilon} \right] \quad (5)$$

where Green functions are defined by $G^\pm(\epsilon) = (\epsilon - H \pm i\delta)^{-1}$, $A = i(G^+ - G^-)$, and $\mathbf{v} = (i/\hbar)[H, \mathbf{r}]$ is the velocity operator. Calculating the trace with eigenfunction (3) and (4) we obtain the following expressions for the diagonal and off-diagonal parts of the conductivity

$$\sigma_{xx} = -\frac{e^3 v_F^2 B}{16\pi^2} \sum_{\alpha\alpha'} \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} d\epsilon f'(\epsilon) A_{n+1\alpha}(\epsilon) A_{n\alpha'}(\epsilon), \quad (6)$$

$$\sigma_{xy} = \frac{e^3 v_F^2 B}{8\pi^2} \sum_{\alpha\alpha'} \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} d\epsilon f(\epsilon) \times \left[\frac{d \text{Re } G_{n+1\alpha}(\epsilon)}{d\epsilon} A_{n\alpha'}(\epsilon) - A_{n+1\alpha}(\epsilon) \frac{d \text{Re } G_{n\alpha'}(\epsilon)}{d\epsilon} \right] \quad (7)$$

where

$$\text{Re } G_{n\alpha}(\epsilon) = \frac{\epsilon - E_{n\alpha}}{(\epsilon - E_{n\alpha})^2 + \Gamma^2}, \quad (8)$$

$$A_{n\alpha}(\epsilon) = \frac{2\Gamma}{(\epsilon - E_{n\alpha})^2 + \Gamma^2}. \quad (9)$$

Here, in order to take into account the impurity scattering, we introduce a damping parameter Γ .

In the low field limit $\hbar\omega_c \ll k_B T$, we can replace the summation over Landau levels in Eqs. (6) and (7) by an integration over continuous variable E . After performing the integration over E and using the Mott's relation (1), in the leading order in magnetic field, we obtain longitudinal and transversal components of the thermopower in

terms of universal functions of $\hbar\omega_c/\Gamma$, $k_B T/\Gamma$, and μ/Γ

$$S_{xx} = -\frac{k_B}{e} \frac{\tilde{K}_{xx}^0}{K_{xx}^0}, \quad (10)$$

$$S_{xy} = \frac{k_B}{e} \left(\frac{\hbar\omega_c}{2\Gamma} \right)^2 \frac{K_{xx}^0 \tilde{K}_{xy}^0 - K_{xy}^0 \tilde{K}_{xx}^0}{(K_{xx}^0)^2} \quad (11)$$

where

$$K_{ij}^0 = \int_{-\infty}^{\infty} \frac{dx}{\cosh^2 \frac{1}{2}x} \Phi_{ij} \left(\frac{k_B T}{\Gamma} x + \frac{\mu}{\Gamma} \right), \quad (12)$$

$$\tilde{K}_{ij}^0 = \int_{-\infty}^{\infty} dx \frac{x}{\cosh^2 \frac{1}{2}x} \Phi_{ij} \left(\frac{k_B T}{\Gamma} x + \frac{\mu}{\Gamma} \right). \quad (13)$$

These formulae are in an agreement with the results obtained previously using slightly different approaches.^{10,12} The universal functions Φ_{ij} are the same as obtained previously for the case of longitudinal and Hall conductivities calculations^{19–21}

$$\Phi_{xx}(x) = 1 + \left(x + \frac{1}{x} \right) \tan^{-1} x, \quad (14)$$

$$\Phi_{xy}(x) = \frac{1}{x} \left(\frac{8x^2}{3(1+x^2)^2} + \frac{1+x^2}{x} \tan^{-1} x - \frac{1-x^2}{1+x^2} \right). \quad (15)$$

From the Eqs. (14) and (15), using the expansion $\Phi_{xx} \approx 2$, $\Phi_{xy} \approx 16x/3$ ($x \ll 1$) and $\Phi_{xx} \approx (\pi/2)|x|$, $\Phi_{xy} \approx (\pi/2) \text{sgn } x$ ($x \gg 1$), the following asymptotic behaviour for the Nernst coefficient at $\mu = 0$ can be obtained.^{10,12}

$$S_{xy} = \frac{2\pi^2 k_B^2 T (\hbar\omega_c)^2}{9e\Gamma^3}, \quad \text{for } k_B T \ll \Gamma. \quad (16)$$

and

$$S_{xy} = \frac{(\hbar\omega_c)^2}{4e\Gamma T}, \quad \text{for } k_B T \gg \Gamma. \quad (17)$$

On the other hand, in the quantum limit where Landau levels are well separated $\hbar\omega_c \gg \max\{k_B T, \Gamma\}$, one needs to evaluate Eqs. (6) and (7) numerically, except for the case when $\Gamma \ll k_B T$. In this case one can approximate the Lorentzian in Eq. (9) by a δ -function. Applying this approximation to Eqs. (6) and (7) and using the Mott's formula (1) we obtain the following analytic results for thermoelectric coefficients

$$S_{xx} = -\frac{k_B}{e} \frac{K_{xy} \tilde{K}_{xy} - (\Gamma/k_B T)^2 K_{xx} \tilde{K}_{xx}}{K_{xy}^2 + (\Gamma/k_B T)^2 K_{xx}^2}, \quad (18)$$

$$S_{xy} = \frac{\Gamma}{eT} \frac{K_{xx} \tilde{K}_{xy} + K_{xy} \tilde{K}_{xx}}{K_{xy}^2 + (\Gamma/k_B T)^2 K_{xx}^2} \quad (19)$$

where functions of $\varepsilon_{n\alpha} = E_{n\alpha}/(2k_B T)$ and $x = \mu/(2k_B T)$ are introduced

$$K_{xx} = \frac{1}{4} \text{sech}^2 x + \frac{1}{2} \sum_{n=1}^{\infty} \sum_{\alpha} n \text{sech}^2(x - \varepsilon_{n\alpha}), \quad (20)$$

$$\tilde{K}_{xx} = \frac{1}{2} \frac{x}{\cosh^2 x} + \sum_{n=1}^{\infty} \sum_{\alpha} \frac{n(x - \varepsilon_{n\alpha})}{\cosh^2(x - \varepsilon_{n\alpha})}, \quad (21)$$

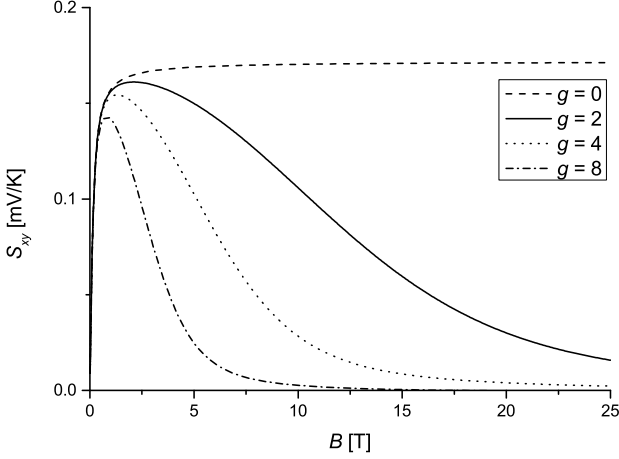


Fig. 1. Magnetic field dependence of the Nernst coefficient at $\mu = 0$ for $T = 1.5$ K, $v_F = 0.5 \times 10^5$ m/s, and $\Gamma/k_B = 3.75$ K for different values of g -factor.

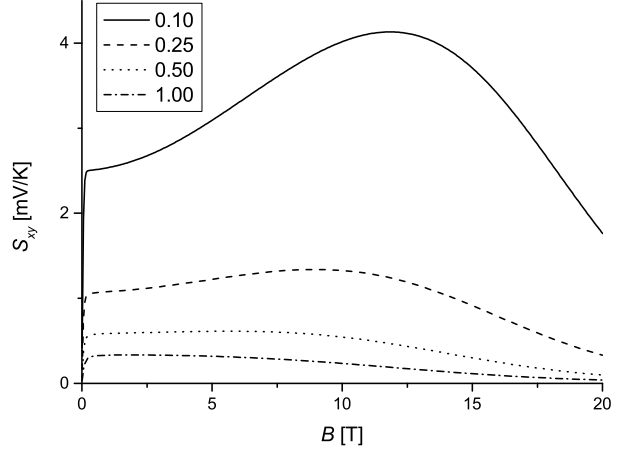


Fig. 2. Magnetic field dependence of the Nernst coefficient at $\mu = 0$ for $T = 1.5$ K, $v_F = 0.5 \times 10^5$ m/s, and $g = 2$ for different values of $\Gamma/(k_B T)$.

$$K_{xy} = \frac{1}{2} \tanh x + \frac{1}{4} \sum_{n=1}^{\infty} \sum_{\alpha} \frac{\sinh 2x}{\cosh(x - \varepsilon_{n\alpha}) \cosh(x + \varepsilon_{n\alpha})}, \quad (22)$$

$$\tilde{K}_{xy} = \varphi(x) + \sum_{n=1}^{\infty} \sum_{\alpha} \varphi(x - \varepsilon_{n\alpha}), \quad (23)$$

and $\varphi(z) = \log(2 \cosh z) - z \tanh z$. The corrections to K_{xy} due to the impurity scattering are of order $\Gamma/(\hbar\omega_c)$,²² and can be omitted.

It is important that, Eqs. (18) and (19) interpolate two typical cases: (I) when μ is away from the Dirac point and (II) when μ is close to the Dirac point ($\mu \approx 0$). In the former case, one can safely neglect the terms of order $(\Gamma/k_B T)^2$, and obtain the results similar to the case of non-relativistic two-dimensional electron gas^{14, 15}

$$S_{xx} = -\frac{k_B}{e} \frac{\tilde{K}_{xy}}{K_{xy}}, \quad (24)$$

$$S_{xy} = \frac{\Gamma}{eT} \frac{K_{xx} \tilde{K}_{xy} + K_{xy} \tilde{K}_{xx}}{K_{xy}^2}. \quad (25)$$

Here the thermopower has a sequence of peaks near the Landau levels. At low temperatures at each Landau level S_{xx} has a universal value $-\text{sgn } n(k_B/e) \log 2/n$, while the Nernst coefficient in this region is small in comparison with S_{xx} .

In the latter case ($\mu \approx 0$), the behaviour of the thermoelectric coefficients changes significantly since K_{xy} vanishes in the vicinity of $n = 0$ Landau level. As a result, the Nernst coefficient has a large peak at $\mu = 0$ with the value given by

$$S_{xy} = \frac{k_B^2 T}{e\Gamma} \frac{\tilde{K}_{xy}}{K_{xx}}. \quad (26)$$

At low T the peak saturates at the value $4k_B^2 T \log 2/(e\Gamma)$, while S_{xx} vanishes. In this region, for non-zero but small μ , the Nernst coefficient can considerably exceed the Seebeck coefficient

In order to describe the behaviour of the thermoelectric coefficients in high magnetic fields we also take into account Zeeman splitting of Landau levels $E_{n\alpha} \rightarrow E_{n\alpha} \pm \Delta$ where $\Delta = (1/2)g\mu_B B$, μ_B and g are the Bohr magneton and g -factor respectively. In magnetic field up to 10 T the Zeeman splitting is small compared with $\hbar\omega_c$. For $v_F = 0.5 \times 10^5$ m/s, $g = 2$, and $B = 1$ T the ratio $\Delta/\hbar\omega_c \approx 0.03$. The resulting magnetic field dependencies of the Nernst coefficient at $\mu = 0$ for $T = 1.5$ K and $v_F = 0.5 \times 10^5$ m/s, and $\Gamma/k_B = 3.75$ K for different values of g are shown in Fig. 1. To obtain Fig. 1 we perform the numerical summation over Landau levels in Eqs. (6), (7) and use the Mott's formula (1). The value $\Gamma/k_B = 3.75$ K is chosen similar to that evaluated previously from the magnetoresistance calculations for α -(BEDT-TTF)₂I₃..²³ In high magnetic field $\hbar\omega_c \gg k_B T$, the Nernst coefficient in the case without Zeeman splitting ($g = 0$) saturates as qualitatively predicted by Eq. (26). This saturating behaviour changes to a decay when Zeeman splitting becomes the same order as temperature and impurity broadening. In low magnetic field the asymptotic behaviour of S_{xy} is described by Eqs. (16) and (17).

The magnetic field dependence of the Nernst coefficient in the presence of Zeeman splitting is different in the (a) $\Gamma \ll k_B T$ and (b) $\Gamma \geq k_B T$ limits which reflects the different mechanisms of Landau level broadening. Figure 2 shows the magnetic field dependence of the Nernst coefficient for several values of $\Gamma/(k_B T)$. In the case (b) with large $\Gamma/(k_B T)$ (dot-dash line in Fig. 2), S_{xy} decreases monotonically except for the very vicinity of $B = 0$. In contrast, in the case (a) with small $\Gamma/(k_B T)$ (solid line in Fig. 2), there is a region in which S_{xy} increases. In this case the increase the Nernst coefficient is understood from Eqs. (20), (23), and (26). Actually, the asymptotic behaviour for large $\Delta/(k_B T)$ is given by

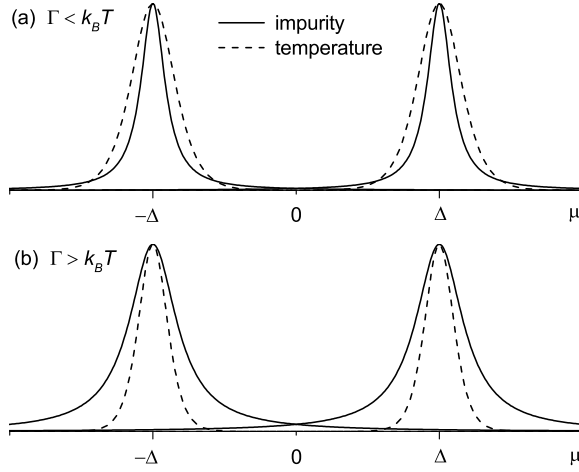


Fig. 3. The difference between the Landau level broadening in (a) $\Gamma < k_B T$ and (b) $\Gamma > k_B T$ case. The impurity (solid) and temperature (dashed) broadenings are given by Lorentzian $\Gamma^2 / [(\mu \pm \Delta)^2 + \Gamma^2]$ and $\text{sech}^2 [(\mu \pm \Delta)/2k_B T]$ respectively.

$$S_{xy} = \frac{k_B^2 T}{e\Gamma} \left[1 + \frac{\Delta}{k_B T} + \left(\frac{3}{2} + \frac{\Delta}{k_B T} \right) e^{-\Delta/(k_B T)} \right] + O(e^{-2\Delta/(k_B T)}). \quad (27)$$

However, the Nernst coefficient starts to decrease in the large B region. This is understood as follows. For sufficiently large Δ the impurity broadening at $\mu = 0$, given by Lorentzian, becomes dominant over the temperature broadening which has exponential decay, as illustrated in Fig. 3 (a). This effect causes the decay of the Nernst coefficient in the large B region where the contribution at $\mu = 0$ comes from the overlap of the split $n = 0$ Landau level. In case (b), the impurity broadening is always dominant over temperature broadening, as illustrated in Fig. 3 (b), and behaviour of the Nernst coefficient is similar to that shown in Fig. 1 and by dash-dot line in Fig. 2.

The temperature dependence of the Nernst coefficient at $\mu = 0$ for $v_F = 0.5 \times 10^5$ m/s, $\Gamma/k_B = 3.75$ K, and $g = 2$, calculated from Eqs. (1), (6) and (7), is shown in Fig. 4 for several values of magnetic field. For large magnetic fields, S_{xy} shows activation behaviour at low temperature due to the Zeeman splitting of $n = 0$ Landau level, while for small B the temperature dependence at low temperature is approximately linear as predicted by Eq. (26). The position of the peak corresponds to the temperature when different Landau levels start to overlap which separates the quantum limit ($\hbar\omega_c \gg k_B T$) from the low field limit ($\hbar\omega_c \ll k_B T$). In the latter case the asymptotic behaviour at high temperatures is given by Eq. (17).

In summary, we have calculated longitudinal and transverse components of the thermopower in quantum limit. For the Nernst coefficient we have calculated the magnetic field and temperature dependencies at $\mu = 0$. These results with the Zeeman term are qualitatively consistent with the recent experiments in α -(BEDT-TTF)₂I₃ organic conductor,⁹ although there are some quantitative discrepancies. First, for the g factor of $g = 2$

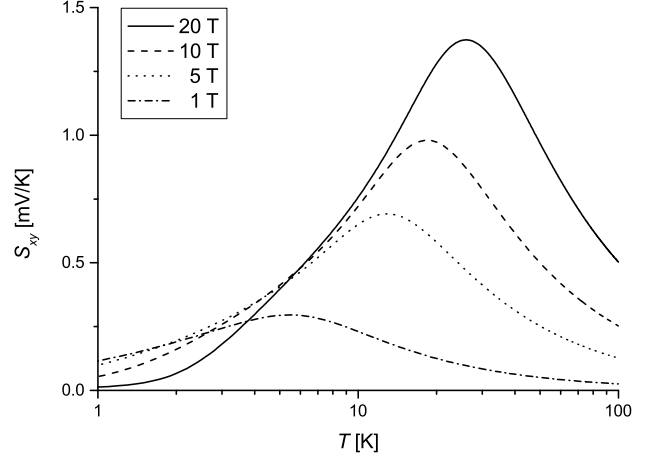


Fig. 4. Temperature dependence of the Nernst coefficient at $\mu = 0$ for $v_F = 0.5 \times 10^5$ m/s and $\Gamma/k_B = 3.75$ K for different values of magnetic field B .

the decay rate of S_{xy} in Fig. 1 as a function of the magnetic field is about a factor 2 smaller than that of experiment. To reproduce the experimental decay rate in our theory, we need to assume $g \approx 6$, which is similar to the effective g -factor discussed in Ref.²⁴ Second, the positions of the peaks on the temperature dependencies shown in Fig. 4 are shifted to higher temperatures than in the experiment. The origin of this shift, as pointed out in Ref.,²⁵ may arise from magnetic field dependence of Γ due to the presence of charged impurities.^{19,22} In order to achieve better agreement between the theory and experiments in α -(BEDT-TTF)₂I₃, it will be necessary to take into account the tilting of the Dirac cone and to use more realistic model for impurity scattering. This remains as a future problem.

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